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Uranium-Series Constraints on Subrepository Water Flow at Yucca Mountain, Nevada

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Abstract — Mineral abundances and whole-rock chemical and uranium-series isotopic compositions were measured in unfractured and rubble core samples from borehole USW SD-9 in the same layers of variably zeolitized tuffs that underlie the proposed nuclear waste repository at Yucca Mountain, Nevada. Uranium concentrations and isotopic compositions also were measured in pore water from core samples from the same rock units and rock leachates representing loosely bound U adsorbed on mineral surfaces or contained in readily soluble secondary minerals. The chemical and isotopic data were used to evaluate differences in water-rock interaction between fractured and unfractured rock and between fracture surfaces and rock matrix. Samples of unfractured and rubble (fragments about 1 centimeter) core and material from fracture surfaces show similar amounts of uranium-series disequilibrium, recording a complex history of sorption and loss of uranium over the past 1 million years. The data indicate that fractures in zeolitized tuffs may not have had greater amounts of water-rock interaction than the rock matrix. The data also show that rock matrix from subrepository units is capable of scavenging uranium with elevated uranium-234/uranium-238 from percolating water and that retardation of radionuclides and dose reduction may be greater than currently credited to this aspect of the natural barrier. Uranium concentrations of pore water and the rock leachates are used to estimate long-term in situ uranium partition coefficient values greater than 7 milliliters per gram.

I. INTRODUCTION

Retardation of radionuclides by sorption on minerals in the rocks along downgradient ground-water flow paths is a positive attribute of the natural barrier at Yucca Mountain, Nevada, the site of a proposed high-level nuclear waste repository. Low-temperature diagenetic alteration of volcanic glass in nonwelded tuffs beneath the proposed repository horizon produced thick, widespread zones of silicified, zeolite-rich rocks with high sorptive capacities. Although both glass and zeolites have similar grain densities, the porosity of the unaltered vitric tuff (30 to 40 percent) is larger than that of the zeolitized and silicified vitric tuff (10 to 30 percent) [1], indicating that glass alteration to zeolites and silica (opal) was accompanied by expansion, reducing the primary void space. As a result of the reduction in matrix porosity, the overall effectiveness of radionuclide retardation by sorption on the zeolitized matrix may be decreased relative to unaltered vitric tuff because water would flow mainly through fractures. However, if an appreciable fraction of total flow continues to occur through the rock matrix in zeolitized units, repository performance will be

enhanced by reducing the radionuclide transport to the downgradient environment.

Isotope ratios in the decay chain of uranium-238 (^{238}U) are indicators of water-rock interaction. In systems older than about 1 million years that remain closed to mass transfer, decay products of ^{238}U reach a state of secular radioactive equilibrium where $^{234}\text{U}/^{238}\text{U}$ and thorium-230 (^{230}Th)/ ^{238}U activity ratios (AR) are unity. However, water-rock interaction along flow paths may result in differential isotope mobility and radioactive disequilibrium in both the water and the rock, the degree of which depends on water velocity, rock dissolution rates, alpha (α)-recoil processes, adsorption and desorption, and the presence of secondary minerals [2]. Compared to matrix flow in the unaltered tuffs, the decreased porosity (and presumably permeability) of the zeolitized and silicified rock matrix would reduce water flux through the matrix resulting in reduced opportunity for isotope exchange, and $^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ AR that are closer to secular equilibrium in unfractured rock samples. The purpose of this paper is to describe long-term water-rock interaction in samples of unfractured and fractured Miocene-age zeolitic tuffs of the Calico Hills

Formation and the Prow Pass Tuff from borehole USW SD-9 using chemical and U-series isotopic indicators of ground-water flow and to assess whether subrepository water flow is dominated by fracture or matrix pathways.

II. WORK DESCRIPTION

Borehole USW SD-9 (total depth of 677.8 meters [m]) is near the northern part of the proposed repository area at Yucca Mountain, west of the main drift of the Exploratory Studies Facility (ESF) (Fig. 1) [3, p. 7]. The regional water table in USW SD-9 is at a depth of 572.3 m [3, p. 7]. Subrepository geologic units investigated in this study include extensively zeolitized and silicified tuffs of the Calico Hills Formation (Tac), which are present in this borehole below a depth of 451.0 m, and the partially zeolitized Prow Pass Tuff (Tep), beginning at a depth of 554.9 m [3, p. 10].

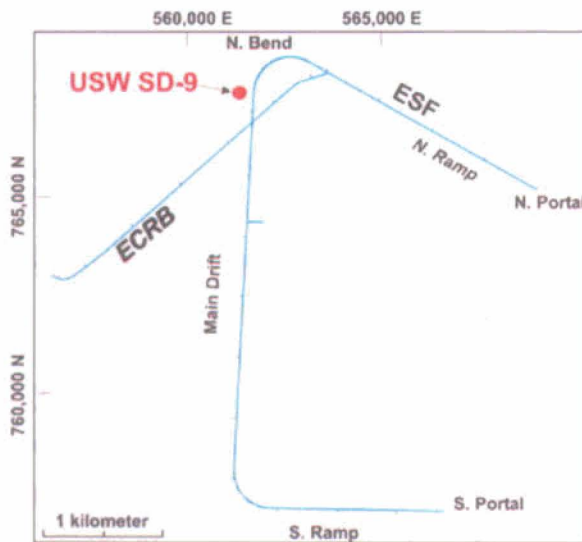


Fig. 1. Location of borehole USW SD-9 relative to the Exploratory Studies Facility (ESF) and Enhanced Characterization of the Repository Block (ECRB) Cross Drift tunnels, Yucca Mountain, Nevada (after Engstrom and Rautman [3]).

Mineral abundances and whole-rock chemical and U-series isotopic compositions were determined in both unfractured core samples representing rock matrix and rubble rock fragments (about 1 centimeter [cm] in diameter) representing zones of higher permeability in the Calico Hills Formation and Prow Pass Tuff in borehole USW SD-9 (sampled depth interval from 451.1 to 633.7 m). Chemical and U-series isotopic compositions also were measured in samples of rock powders collected using a dental burr from surfaces of natural fractures in core from the same depth interval. These samples

represent potential fracture pathways in the Calico Hills Formation and Prow Pass Tuff. The analyses were used to evaluate possible differences in water-rock interaction in fracture and matrix flow. In addition, U concentrations and isotopic compositions were measured in samples of pore water obtained by ultracentrifugation (spun pore water) of unfractured USW SD-9 core samples or in pore water salts obtained by leaching rubble samples with deionized water. The concentrations and isotopic compositions of loosely bound U contained in carbonates or adsorbed on reactive mineral surfaces of the same rocks pre-leached with deionized (DI) water were obtained by analyzing leachates using 1 molar (M) sodium acetate (NaOAc) solution adjusted to pH 5 with acetic acid. Uranium in readily acid-soluble secondary minerals was further characterized using 8-normal (N) nitric acid (HNO₃) leachates of splits of samples pre-leached with deionized water.

Mineral abundances were measured by quantitative X-ray powder diffraction methods (QXRD) using the full-pattern analysis program FULLPAT [4]. Chemical compositions of major and trace elements were determined by several techniques including standard X-ray fluorescence spectroscopy (XRF) and inductively coupled plasma-mass spectrometry (ICP-MS) techniques. Uranium and Th isotopes were measured using a Triton Thermo Finnigan® thermal ionization mass spectrometer (TIMS) equipped with a retarding potential quadrupole (RPQ). Compositions of international isotope standards (NIST SRM-4321 and IRMM-036) were used to evaluate the accuracy and precision of isotope measurements: replicate analyses of ²³⁴U/²³⁵U (0.007291±0.000009 [2 sigma [σ], n=99]) for NIST SRM-4321, and of ²³²Th/²³⁰Th (321,500±3,000 [2σ, n=26] for IRMM-036) are within analytical uncertainty of certified values. Routinely measured U and Th total procedure blanks contribute less than 0.02 percent of U and Th in the samples. Typical within-run relative 2σ uncertainties of U and Th isotopic ratios of unknowns were about 0.2 and 0.5 percent, respectively. Replicate analyses of a powdered welded tuff from borehole USW SD-9 (depth 283.6 m) by isotope dilution TIMS yielded a mean U concentration of 4.52±0.03 micrograms per gram (μg/g) (2σ, n=16), Th concentration of 23.4±0.2 μg/g (2σ, n=6), ²³⁴U/²³⁸U AR of 0.987±0.002 (2σ, n=16) and ²³⁰Th/²³⁸U AR of 0.985±0.026 (2σ, n=6). Atomic ratios were converted to activity ratios using known decay constants [5, 6].

III. RESULTS

The dominant secondary mineral in unfractured and rubble samples of zeolitized and silicified tuff (n=22) is clinoptilolite (37.3 to 73.0 percent, found in 19 samples, Table 1) with lesser amounts of opal-CT (6.6 to 20.8

percent, found in 19 samples) and mordenite (1.2 to 22.4 percent, found in 17 samples). Scanning electron microscope images of zeolite minerals on fracture surfaces in core samples show tabular to fibrous mineral forms (Fig. 2). Smectite is detected in all samples ranging from 0.1 to 17.8 percent. Devitrified tuffs are present in

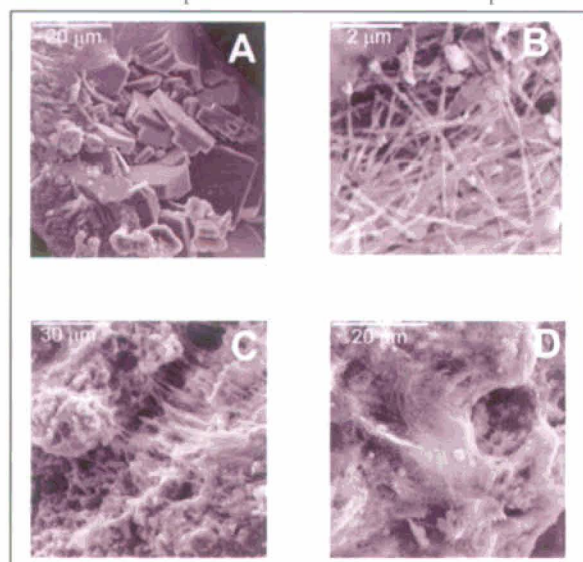


Fig. 2. Scanning electron microscope images of zeolite minerals on natural fracture surfaces in core samples from borehole USW SD-9, Yucca Mountain, Nevada: (A) tabular clinoptilolite from 451.1 m depth (Calico Hills Formation, Tac); (B) fibrous mordenite from 474.8 m (Tac); (C) fibrous mordenite from 481.7 m (Tac); and (D) fibrous mordenite from 544.6 m (Calico Hills Formation bedded tuff, Tacbt).

the Tcp at depths of 582.1 to 613.4 m [3]. Three samples collected within this interval have about equal amounts of quartz, potassium feldspar, and plagioclase and lack zeolites (Table 1).

Lithology-dependent chemical variations in unfractured ($n=15$) and rubble ($n=7$) borehole USW SD-9 samples are shown with depth in Fig. 3. The largest variations in concentrations are in the bedded tuff (Tacbt) and basal tuffaceous sandstone (Tacbs) samples from the lower portion of the Calico Hills Formation and the samples from the unsaturated zone (UZ) portion of the Prow Pass Tuff (Fig. 3). Decreases in calcium (Ca) concentrations and increases in sodium (Na) concentrations with depth through the upper 50 m of the Tac (Fig. 4) are consistent with published results documenting accumulation of Ca accommodated by loss of Na as a result of downward water movement and cation exchange within the zeolite sequence [7]. However, systematic variations with depth and zeolite abundance are not observed for U and Th concentrations over this same interval. Samples of unfractured and rubble core

have similar mean concentrations of U (4.64 ± 0.99 [1σ] and 4.38 ± 0.76 [1σ] $\mu\text{g/g}$, respectively) and Th (20.5 ± 2.4 and 20.8 ± 1.0 $\mu\text{g/g}$,

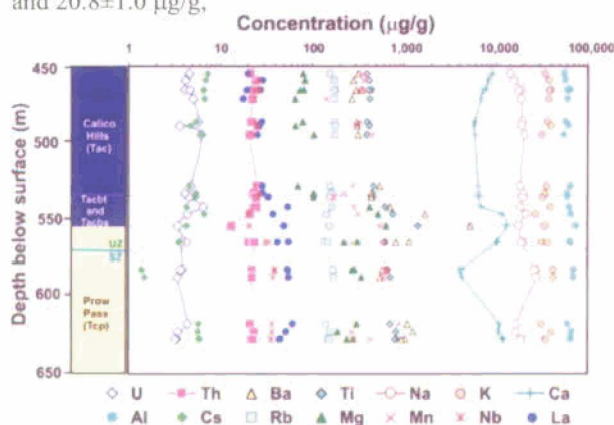


Fig. 3. Selected element concentrations with depth in unfractured and rubble whole-rock samples from borehole USW SD-9 drill core, Yucca Mountain, Nevada. Depths for lithostratigraphic unit boundaries and water table are from Engstrom and Rautman [3]. UZ and SZ are unsaturated and saturated zones, respectively.

respectively). Samples of rock powders collected from fracture surfaces ($n=13$) show larger variability in U and Th concentrations and on average are enriched in both elements (6.7 ± 3.9 [1σ] $\mu\text{g/g}$ U and 25.5 ± 5.2 [1σ] $\mu\text{g/g}$ Th).

Samples of unfractured core, rubble core, and rock powders collected from fracture surfaces have similar $^{234}\text{U}/^{238}\text{U}$ AR ranging from 0.93 to 1.16, from 0.98 to 1.12, and from 0.92 to 1.15, respectively, indicating both enrichments ($^{234}\text{U}/^{238}\text{U}$ AR > 1) and depletions ($^{234}\text{U}/^{238}\text{U}$ AR < 1) in the daughter ^{234}U relative to the parent ^{238}U (Fig. 5A). In contrast to the rock samples, all pore water and rock leachate samples have elevated $^{234}\text{U}/^{238}\text{U}$ AR ranging from 1.1 to 5.2 (Fig. 5B).

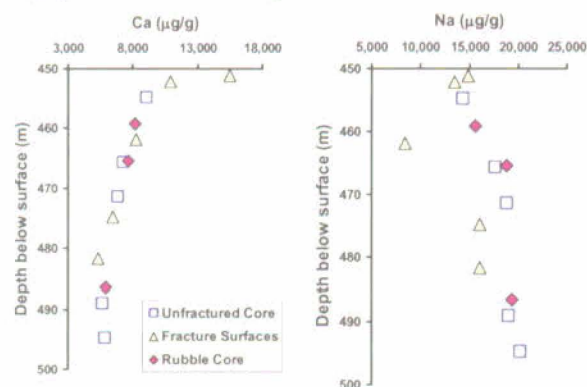


Fig. 4. Calcium (Ca) and sodium (Na) concentrations with depth for whole-rock samples of borehole

USW SD-9 drill core, Yucca Mountain, Nevada, from the upper 50 meters of the Calico Hills Formation.

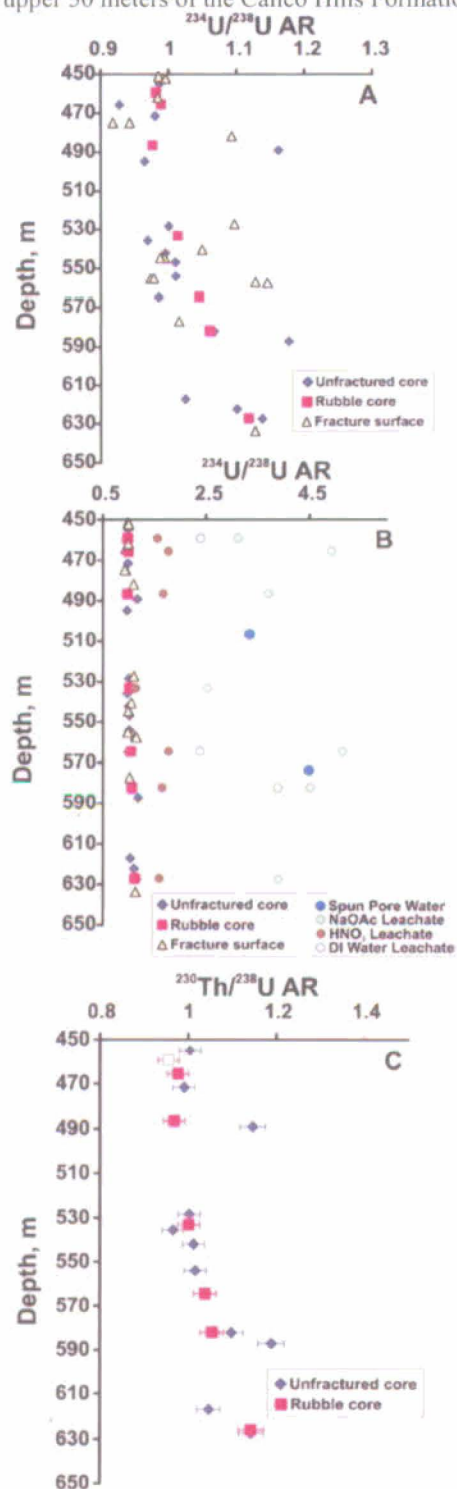


Fig. 5. Activity ratios (AR) with depth in whole-rock, pore water, and leachate analyses of borehole USW SD-9 drill core, Yucca Mountain, Nevada: (A) whole-rock

$^{234}\text{U}/^{238}\text{U}$ AR for samples of unfractured core, rubble core, and fracture surfaces; (B) $^{234}\text{U}/^{238}\text{U}$ AR data for pore water and rock leachates with whole-rock data from (A) shown for comparison (note difference in X-axis scales between A and B); and (C) whole-rock $^{230}\text{Th}/^{238}\text{U}$ AR in samples of unfractured and rubble core. Error bars (2σ) are shown when they are larger than symbol sizes.

Unfractured and rubble core samples also have overlapping $^{230}\text{Th}/^{238}\text{U}$ AR values (Fig. 5C) that are similar to their values of $^{234}\text{U}/^{238}\text{U}$ AR. Data plot close to the $^{234}\text{U}/^{238}\text{U}$ - $^{230}\text{Th}/^{238}\text{U}$ equiline in Fig. 6, indicating that ^{230}Th and ^{234}U activities are essentially equal. The amount of ^{234}U disequilibrium in rock powders from fracture surfaces ($^{234}\text{U}/^{238}\text{U}$ AR 0.92 to 1.15) is similar to unfractured core. No correlations are observed between zeolite (clinoptilolite and mordenite) abundance and the degree of U-series disequilibrium, although $^{234}\text{U}/^{238}\text{U}$ AR and $^{230}\text{Th}/^{238}\text{U}$ AR increase slightly with depth (Figs. 5A and 5C).

Uranium concentrations in NaOAc leachates (0.012 to 0.071 $\mu\text{g/g}$ rock) are lower than those in HNO₃ leachates (0.47 to 1.38 $\mu\text{g/g}$ rock). Comparisons with whole-rock data indicate that NaOAc leaching released 0.3 to 1.7 percent of total rock ^{238}U and HNO₃ leaching released 10.1 to 32.7 %. Percentages for leached ^{234}U are higher and range from 1.1 to 5.5% for NaOAc leachates and from 15.7 to 46.1% for HNO₃ leachates. These data allowed estimates of time-integrated *in situ* U partition coefficients ($K_d = C_s/C_w$, where C_s and C_w represent concentrations measured in the solid and water, respectively) under natural flow conditions. Because U was present in these rocks at the time of their formation, a large fraction of C_s is tightly bound within mineral lattices. Therefore, the partition coefficient calculated using whole-rock C_s grossly overestimates the value for K_d [8]. For instance, use of median U concentrations in pore water (5 nanograms per milliliter [ng/mL]) and in rocks (4.5 $\mu\text{g/g}$) yields an upper limit of the ^{238}U K_d value of about 900 milliliters per gram [mL/g]. Substantially smaller ^{238}U K_d values of about 7 and 180 mL/g are obtained using median C_s values of 0.035 and 0.90 $\mu\text{g/g}$ measured in NaOAc and HNO₃ rock leachates, respectively. These values more accurately represent the mobile U component that is adsorbed on mineral surfaces or contained in readily acid-soluble secondary minerals.

IV. DISCUSSION AND CONCLUSIONS

Data for samples of whole rocks, fracture surfaces and pore water collected in this study indicate that the tuffs that underlie the proposed repository record a complex history of both sorption and loss of U in the last million years. However, similar variations in $^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ AR values for unfractured core, rubble core, and fracture surfaces imply that fractures in

zeolitized tuffs did not have greater amounts of water-rock interaction than the rock matrix and, therefore, that water flow may not be dominated by fracture pathways. Decreasing Ca concentrations and increasing Na concentrations with depth through the upper 50 m of the Tac in unfractured samples confirm previous observations [7] that the solute transport into the zeolitized tuffs took place despite the decrease in permeability in the transition from vitric nonwelded to zeolitized rock [1].

Values of whole-rock $^{234}\text{U}/^{238}\text{U}$ AR commonly are greater than 1 in the zeolitized tuff samples analyzed in this study. This result contrasts with data from samples of the welded part of the Topopah Spring Tuff (Tpt), the proposed repository horizon, that invariably has $^{234}\text{U}/^{238}\text{U}$ AR < 1. The depletion in ^{234}U in Tpt rocks is caused by losses through α -recoil processes to water percolating downward through the UZ [9-13], which becomes enriched in ^{234}U . Therefore, the large $^{234}\text{U}/^{238}\text{U}$ AR in pore water U (centrifuged water and deionized water leachates) and in U sorbed on mineral surfaces (NaOAc leachates) observed in this study indicate the presence of uranium transported in water percolating from higher levels within the UZ. In addition, borehole USW SD-9 whole-rock samples from units beneath the proposed repository having $^{234}\text{U}/^{238}\text{U}$ AR > 1 strongly support the potential for sorption of U from percolating water by zeolitic rocks. The high sorptive capacity of these rocks is enhanced by large surface area of tabular to fibrous mineral forms (Fig. 2), which is about 10 times larger in zeolitic tuffs than in devitrified tuffs and about 30 times larger than in vitric tuffs (average values are 28.7, 2.6, and 1.0 square meter per gram (m^2/g), respectively [14]).

Rates of U mobility in subrepository units are constrained by U-series isotope compositions shown in Fig. 6. The fact that ^{234}U and ^{230}Th have similar activities and plot close to the equiline for both unfractured and rubble rock samples indicates that rates of ^{238}U leaching from zeolitized rocks are sufficiently low to allow ^{230}Th to grow back into radioactive equilibrium with its immediate precursor ^{234}U (requiring about 300 thousand years) while leaving ^{234}U in appreciable disequilibrium with its parent ^{238}U (requiring >1 My to restore secular radioactive equilibrium). Preliminary modeling of the U-series data for the ^{234}U -depleted Tac whole-rock samples using steady-state solutions of the continuous U leaching model by Latham and Schwarcz [15] implies that the ^{238}U leaching rate constant was about $10^{-8} \text{ years}^{-1}$ (y^{-1}). Possibly, more rigorous modeling of whole-rock U-series data along with $^{234}\text{U}/^{238}\text{U}$ AR data for pore water from

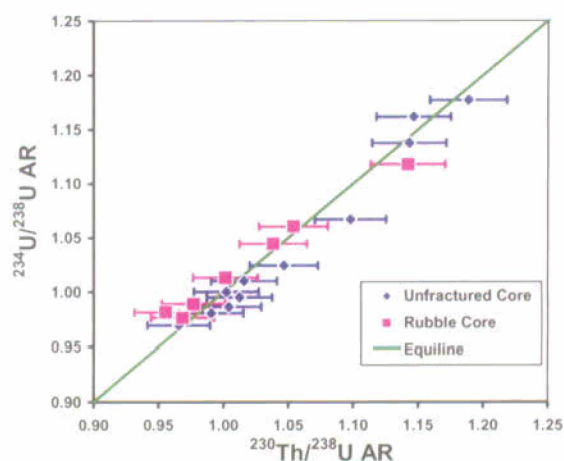


Fig. 6. $^{230}\text{Th}/^{238}\text{U}$ plotted in relation to $^{234}\text{U}/^{238}\text{U}$ activity ratios (AR) in whole-rock samples from borehole USW SD-9 drill core, Yucca Mountain, Nevada. Error bars equal to 2-sigma (σ) uncertainties. Equiline represents the loci of points with equal ^{234}U and ^{230}Th activities ($^{230}\text{Th}/^{234}\text{U}$ AR=1).

borehole USW SD-9 will allow estimation of water flux percolating through the subrepository units.

Measurements of the sorption distribution coefficient, K_d , are critical in the determination of sorption-induced retardation of radionuclide transport. All modeling predictions of radionuclide fate and transport through the unsaturated zone at Yucca Mountain are currently based on K_d values determined by laboratory batch experiments using crushed tuffs [16]. Uranium concentration data for pore water and the rock leachates from core samples of borehole USW SD-9 allow estimation of long-term *in situ* K_d values for U greater than 7 mL/g, which are substantially larger than the value of 0.5 mL/g currently used for zeolitic units at Yucca Mountain ([16], Table 5-1). U-series isotope data from core samples of borehole USW SD-9 show that zeolitized and silicified rocks from subrepository units can scavenge U with $^{234}\text{U}/^{238}\text{U} > 1$ from percolating water and that amounts of retardation of radionuclides may be greater than currently credited to the zeolitized and silicified rocks part of the natural barrier.

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TABLE I. Mineral Abundances in Percent for Core Samples of Tuffs beneath the Proposed Repository Horizon at Yucca Mountain, Nevada, from Borehole USW SD-9 Determined by Quantitative X-Ray Diffraction (QXRD).

Sample	Depth (m)	Stratigraphy*	Smec-tite	Clino-ptilolite	Morde-nite	Cristo-balite	Opal-CT	Quartz	K-Feldspar	Plagio-clase	Hema-tite	Mica	Total
<i>Unfractured Core</i>													
USW SD9-1492.4/1492.6	454.9	Calico Hills Formation (Tac)	1.8**	66.9	n.d.***	n.d.	20.8	3.1	3.7	4.6	n.d.	0.3	101.3
USW SD9-1528.1/1528.3	465.8	"	2.5	69.0	1.8	n.d.	13.8	3.2	4.7	5.3	n.d.	0.4	101.0
USW SD9-1546.9/1547.1	471.5	"	0.4	73.0	5.7	n.d.	12.0	3.1	3.8	3.0	n.d.	0.5	101.5
USW SD9-1604.6/1604.8	489.1	"	2.3	68.4	9.4	n.d.	10.8	1.6	4.0	3.4	n.d.	0.4	100.2
USW SD9-1733.6/1733.8	528.4	"	8.3	54.4	14.9	n.d.	13.4	2.9	6.2	2.4	n.d.	n.d.	102.6
USW SD9-1757.6/1757.8	535.7	"	3.8	43.6	21.6	n.d.	13.6	4.8	7.8	5.8	n.d.	1.0	102.1
USW SD9-1778.8/1779.0	542.2	Bedded tuff (Tacbt)	0.1	59.0	7.2	n.d.	12.3	6.7	10.4	5.7	n.d.	2.0	103.3
USW SD9-1793.8/1794.0	546.8	"	6.2	39.6	22.4	n.d.	6.6	8.0	6.1	11.8	n.d.	2.1	102.9
USW SD9-1818.0/1818.2	554.1	Tuffaceous sandstone (Tacbs)	17.8	38.1	n.d.	n.d.	10.1	5.4	9.4	15.4	n.d.	3.1	99.3
USW SD9-1853.1/1853.3	564.8	Prow Pass Tuff (Tep)	2.4	55.7	6.2	n.d.	12.6	3.7	10.8	10.5	n.d.	0.1	101.9
USW SD9-1910.4/1910.6	582.3	"	3.7	n.d.	n.d.	1.0	n.d.	29.0	32.0	30.5	0.8	1.4	98.4
USW SD9-1926.0/1926.2	587.0	"	3.9	n.d.	n.d.	2.8	n.d.	26.2	31.9	30.0	0.6	1.7	97.3
USW SD9-2024.8/2025.0	617.2	"	5.5	48.1	9.0	n.d.	14.6	3.3	9.2	11.9	n.d.	0.6	102.3
USW SD9-2041.7/2041.9	622.3	"	6.3	37.3	11.9	n.d.	12.2	3.7	13.1	14.1	n.d.	1.0	99.6
USW SD9-2058.2/2058.4	627.3	"	3.9	49.0	12.1	n.d.	7.0	3.3	10.3	13.1	n.d.	1.3	99.9
<i>Rubble Core</i>													
USW SD9-1506.5/1506.7	459.2	Calico Hills Formation (Tac)	0.8	71.7	1.2	2.3	11.4	3.4	4.2	4.4	n.d.	0.1	99.5
USW SD9-1527.1/1527.4	465.5	"	3.3	69.0	3.4	3.2	9.1	2.5	5.3	5.5	n.d.	n.d.	101.3
USW SD9-1596.1/1597.0	486.5	"	1.2	62.6	12.2	2.1	13.4	1.4	2.6	2.2	n.d.	n.d.	98.2
USW SD9-1749.2/1749.7	533.2	"	4.7	48.4	21.2	3.4	7.0	5.3	6.4	4.0	n.d.	0.1	100.4
USW SD9-1852.0/1852.6	564.5	Prow Pass Tuff (Tep)	2.7	58.7	4.7	3.2	7.9	5.4	6.8	10.8	n.d.	0.8	101.2
USW SD9-1909.7/1910.0	582.1	"	2.7	n.d.	n.d.	1.3	n.d.	29.0	30.3	31.7	0.6	2.2	97.7
USW SD9-2057.3/2057.5	627.1	"	3.0	44.0	15.1	3.4	7.2	2.9	9.5	14.2	n.d.	1.2	100.5

* Based on stratigraphic units upper contacts and unit thicknesses from Engstrom and Rautman [3, p. 9].

** Typical error for all mineral abundances is ± 0.1 .

*** n.d.- not detected.